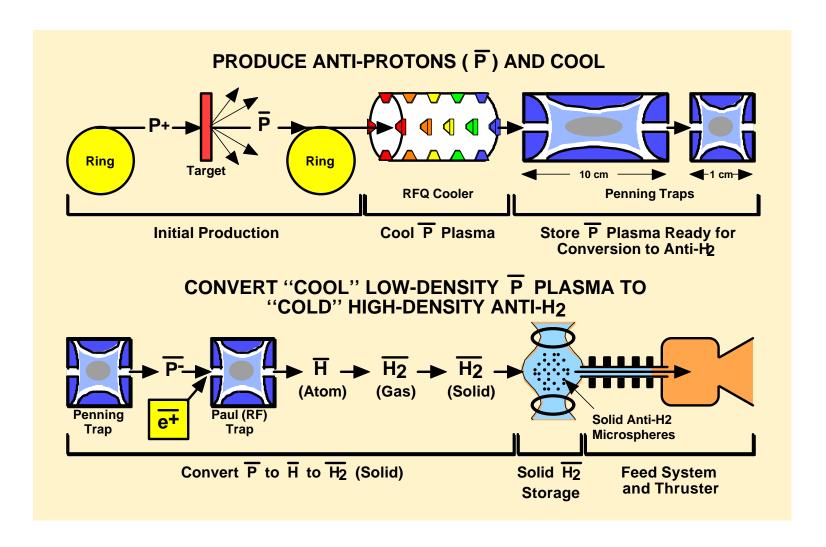
# Photodynamics in Cryogenic Solid Parahydrogen: Application to Bulk Antimatter Storage

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- \* Requirements for Bulk Antimatter Storage
- \* High Energy Density Matter (HEDM) Program
- \* Rapid Vapor Deposition of <u>Transparent</u> Parahydrogen (pH<sub>2</sub>) Solids
- \* CO/pH<sub>2</sub>: a Molecular Thermometer
- \* Proposal: Magnetic Levitation & Vacuum Storage of Solid pH<sub>2</sub> Optical Refrigeration of Solid pH<sub>2</sub> (?!)

#### **Antimatter Production and Storage**



[R.H. Frisbee, Adv. Prop. Concepts Webpage, http://sec353.jpl.nasa.gov/apc/Antimatter/05.html]

#### **Previous/Ongoing Normal Matter Efforts**

- \* Laser stimulated radiative recombination of  $H^+ + e^- \rightarrow H$  [M.L. Rogelstad, F.B. Yousif, T.J. Morgan, and J.B.A. Mitchell, J. Phys. B **30**, 3913 (1997)]
- \* Magneto-optical trapping of H atoms
  [D. Kleppner, et al., Phys. Rev. Lett. 81, 3807 (1998), Phys. Rev. Lett. 81, 3811 (1998)]
- \* Photoassociation of Spin-Polarized Hydrogen  $\rightarrow$  H<sub>2</sub> [A.P. Mosk, M.W. Reynolds, T.W. Hijmans, and J.T.M. Walraven, Phys. Rev. Lett. **82**, 307 (1999)]
- \* Laser cooling & trapping of molecules (proposal)
  [J.T. Bahns, W.C. Stwalley, and P.L. Gould, J. Chem. Phys. **104**, 9689 (1996)]
- \* Clustering of trapped  $H_2$  molecules  $\rightarrow (H_2)_n$ ?
- \* Magnetic levitation of condensed hydrogen [C.G. Paine and G.M. Seidel, Rev. Sci. Instrum. **62**, 3022 (1991)]

# Levitation of Liquid and Solid Hydrogen

#### Magnetic levitation of condensed hydrogen

C. G. Paine and G. M. Seidel Department of Physics, Brown University, Providence, Rhode Island 02912

(Received 19 July 1991; accepted for publication 11 August 1991)

Liquid and solid molecular hydrogen has been levitated using a pair of small superconducting solenoids. The hydrogen samples, up to 3 mm in dimension, were trapped in a magnetic potential having either a discrete minimum or a minimum in the form of a ring 1 cm in diameter. The hydrogen could be moved about in the magnetic trap by applying an electric field.

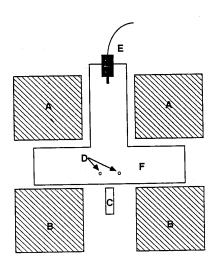


FIG. 1. Cross section of the superconducting magnets and experimental chamber for levitating hydrogen: (A) upper coil; (B) lower coil; (C) NdFeB permanent magnet; (D) location of circular potential energy minimum (see text); (E) injection mechanism for liquid hydrogen; (F) experimental chamber for containing hydrogen and helium gas.

$$d \approx 3 \text{ mm}$$
  
 $m \approx 1 \text{ mg}$ 

$$B(dB/dz) \approx 500 \text{ T}^2/\text{m}$$

$$T \approx 10 - 14 \text{ K}$$
 $P_{H2} \approx 2 - 55 \text{ Torr}$ 
 $\Rightarrow \text{ viscous damping forces}$ 

[C.G. Paine and G.M. Seidel, Rev. Sci. Instrum. 62, 3022 (1991)]

# **Antimatter "Show Stoppers"**

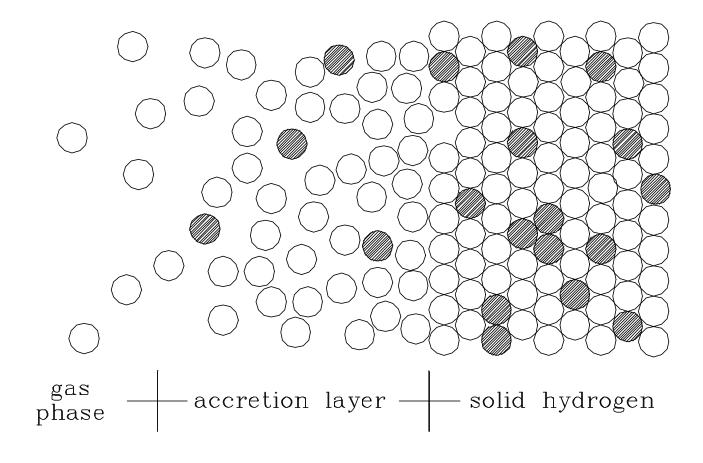
"During the study, some specific technological areas were uncovered where there may be a "show stopper. In assigning priorities, it would be important to make sure that these questionable areas are studied first."

"Antihydrogen ice must be kept below 2 K to keep its vapor pressure low enough so that the antihydrogen molecules sublimating from its surface do not heat up the storage chamber walls. It may be found that radiation cooling to the cold chamber walls is not adequate for extracting heat from the ice generated by unavoidable heat leaks, and no other cooling technique works."

[R.L. Forward, Antiproton Annihilation Propulsion, AFRPL-TR-85-034 (1985)]

#### **HEDM Cryosolid Propellants Concept**

Use cryogenic solid hydrogen as a "packaging material" to store energetic species such as metal atoms and clusters.



#### Dopant Reactions Within Solid H<sub>2</sub>

\* ideally:

$$X + H_2 \qquad \underline{T=2K} \quad X/H_2$$

isolated atoms

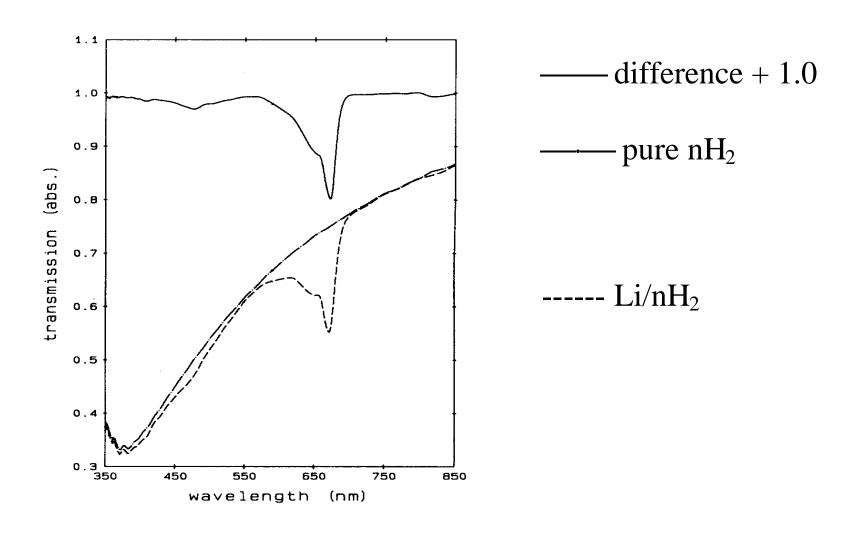
\* in practice:

$$\begin{array}{ccc} X + X + M & \rightarrow & X_2 + M & \text{recombination} \\ & \rightarrow & X_n & \end{array}$$

$$X + H_2 + M \rightarrow HX + H + M$$
 reaction  $\rightarrow H_nX + M$ 

$$\begin{array}{ccc} X_n + H_2 + M & \rightarrow & HX_n + H + M & & both \\ & \rightarrow & H_m X_n + M & & \end{array}$$

#### Transmission Spectrum of Li/nH<sub>2</sub>, d » 10 m



M.E. Fajardo, J. Chem. Phys. **98**, 110 (1993).

# Optical Scattering in Solid Hydrogen

#### **Crystal Growing and Quality (p. 81)**

"There is a considerable art to growing hydrogen crystals of high quality. Good crystals are always grown slowly from the melt; a rapid freeze from the gas produces snow."

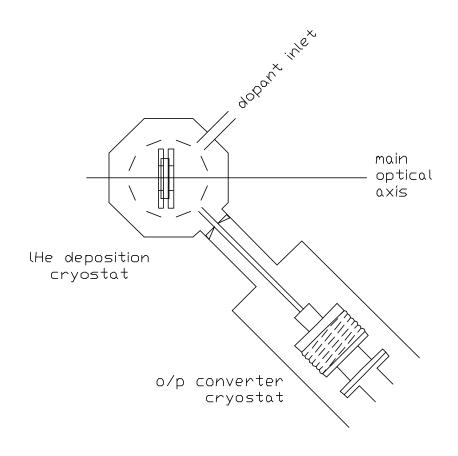
#### Crystallite Light Scattering (p. 83)

"The reason that a good hydrogen crystal is so hard to see is its low refractive index...an estimated 1.16!

Yet a 1 mm-thick layer of hydrogen crystallites can be a completely opaque brown-black."

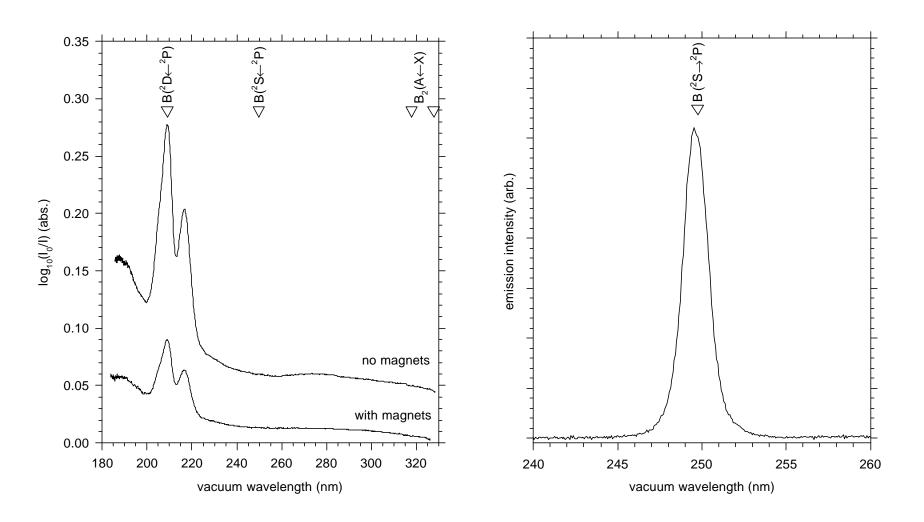
[P.C. Souers, Hydrogen Properties for Fusion Energy (UC Press, Berkeley, 1986)]

# Rapid Vapor Deposition of Gram-Scale Optically Transparent pH<sub>2</sub> Solids (c1997)



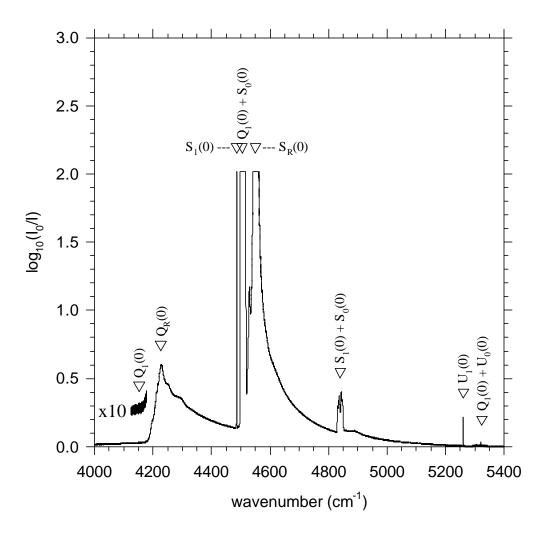
M.E. Fajardo and S. Tam, J. Chem. Phys. **108**, 4237 (1998). S. Tam and M.E. Fajardo, Rev. Sci. Instrum. **70**, 1926 (1999).

### Electronic Spectroscopy of B/pH<sub>2</sub> (d»2 mm)



S. Tam, M. Macler, M.E. DeRose, and M.E. Fajardo, J. Chem. Phys. **113**, 9067 (2000). [J.R. Krumrine, S. Jang, G.A. Voth, and M.H. Alexander, J. Chem. Phys. **113**, 9079 (2000)]

### IR Absorption of 6 mm Thick pH<sub>2</sub> Solid



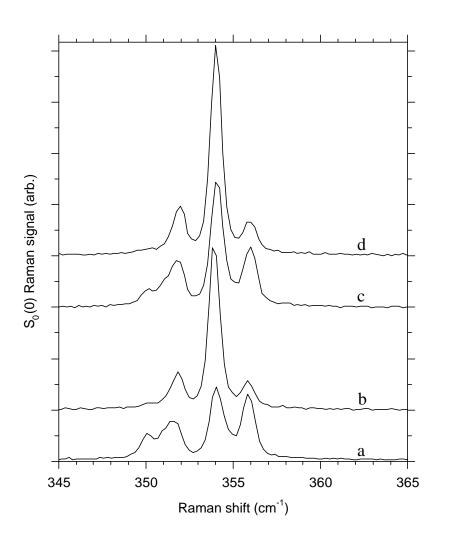
Non-observation of the  $Q_1(0)$  transition demonstrates the absence of  $oH_2$  impurities, and that the microscopic structure is not amorphous or porous.

Observation of  $S_1(0)$  transition demonstrates the absence of inversion symmetry for <u>some</u>  $H_2$  molecular environments.

[van Kranendonk and Gush, Phys. Lett. 1, 22 (1962)]

M.E. Fajardo and S. Tam, J. Chem. Phys. 108, 4237 (1998).

#### Raman Spectra of pH<sub>2</sub> Solids

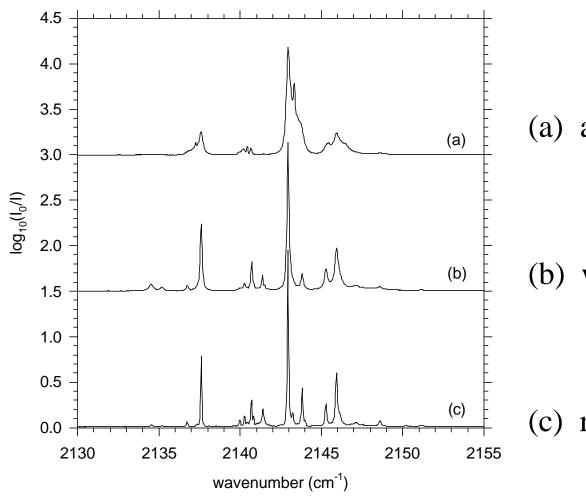


Mixed hcp/fcc as-deposited structure, anneals to hcp. [G.W. Collins, et al., Phys. Rev. B **53**, 102 (1996)]

- (d) sample in (c) warmed to 4.5 K.
- (c) 4.5 mm sample as deposited at 3.3 K ( $\Phi$  = 290 mmol/hr).
- (b) sample in (a) warmed to 4.5 K.
- (a) 6 mm sample as deposited at  $3.1 \text{ K} (\Phi = 200 \text{ mmol/hr}).$

M.E. Fajardo and S. Tam, J. Chem. Phys. 108, 4237 (1998).

# 80 PPM CO/pH<sub>2</sub> (res = $0.1 \text{ cm}^{-1}$ )



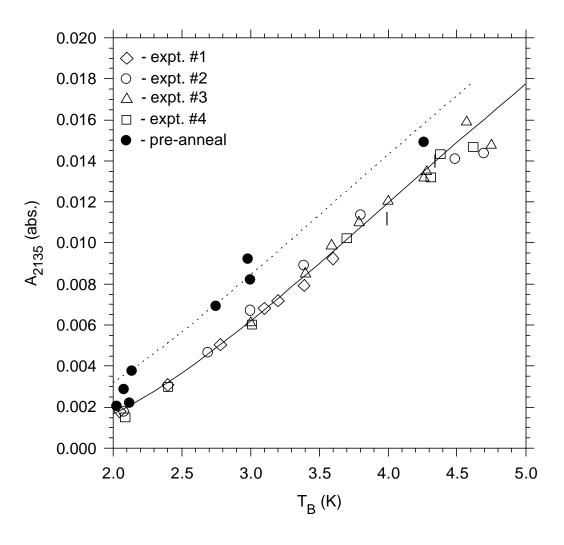
(a) as deposited at T = 2.4 K

(b) warmed to T = 4.8 K

(c) re-cooled to T = 2.4 K

Analysis in collaboration with Prof. T. Momose, Kyoto U.

#### Calibration of CO/pH<sub>2</sub> Thermometer



simplified Boltzmann:

$$A_{2135} = N \exp(-E/T_{CO})$$

$$N = 0.0860$$

$$E = 7.896 \text{ K}$$

include  $Q_{rot} \Rightarrow E \approx 11 \text{ K}$ 

$$6 B_{\text{CO/pH2}} \approx 12 \text{ K}$$

M.E. Fajardo and S. Tam, J. Low Temp. Phys., 122, xxx (2001).

### **HEDM Cryosolids Summary**

- \* Trapped Li, B, Na, Mg, Al atoms in solid hydrogen; c < 0.1%, efforts to achieve higher dopant concentrations are ongoing.
- \* Demonstrated production of doped, gram-scale, optically transparent pH<sub>2</sub> solids by rapid vapor deposition.
- \* Demonstrated spectroscopically that vapor deposited pH<sub>2</sub> solids are densest close-packed solids, NOT amorphous.
- \* Demonstrated suitability of vapor deposited pH<sub>2</sub> solids as hosts for high-resolution ro-vibrational spectroscopy.
- \* Demonstrated CO/pH<sub>2</sub> thermometer diagnostic.
- \* Now apply this technology to bulk antimatter storage!

# Optical Refrigeration in Yb<sup>3+</sup> Doped Glasses

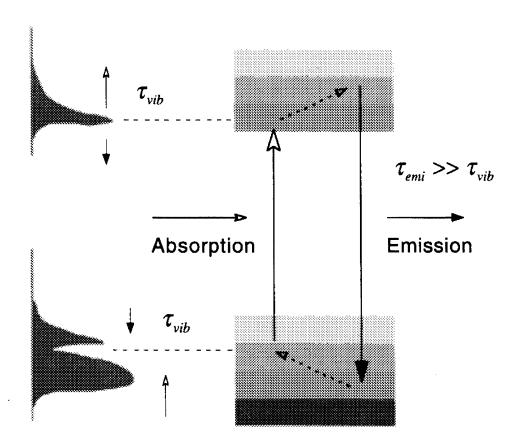


Fig. 1. Schematic illustration of laser-induced fluorescence cooling, where  $\tau_{\rm em}$  is the radiative lifetime and  $\tau_{\rm vib}$  is the phonon relaxation time.

[G. Lei, J.E. Anderson, M.I. Buchwald, B.C. Edwards, R.I. Epstein, M.T. Murtagh, and G.H. Sigel, Jr., IEEE J. Quant. Electr. **34**, 1839 (1998)]

#### **Optical Refrigeration Thermodynamics**

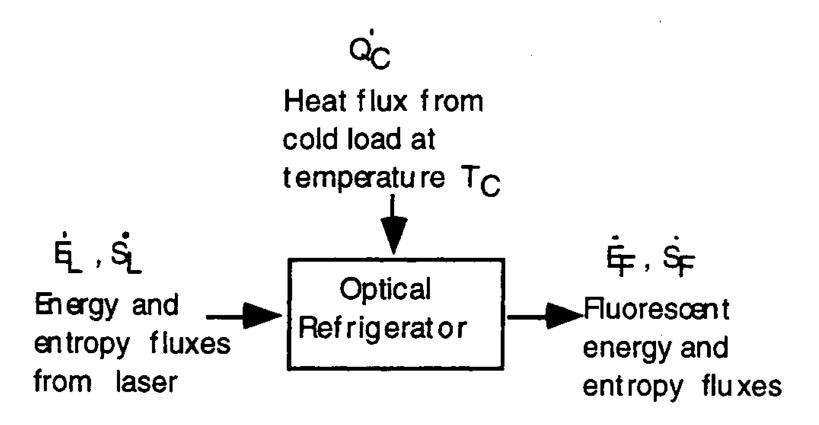


FIG. 7. The energy and entropy fluxes for optical refrigeration.

[B.C. Edwards, M.I. Buchwald, and R.I. Epstein, Rev. Sci. Instrum. 69, 2050 (1998)]

### Nonlinear Optics in Solid pH<sub>2</sub>

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PHYSICAL REVIEW LETTERS

14 July 1997

#### Self-Induced Phase Matching in Parametric Anti-Stokes Stimulated Raman Scattering

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(Received 27 November 1996)

#### $Q_1(0)$ Raman linewidth < 14 MHz (0.0005 cm<sup>-1</sup>) FWHM

PHYSICAL REVIEW A

VOLUME 60, NUMBER 2

**AUGUST 1999** 

#### Subfemtosecond pulse generation with molecular coherence control in stimulated Raman scattering

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#### **Proposed Experimental Efforts**

\* Low Risk:

Magnetic levitation of doped pH<sub>2</sub> solids in high vacuum use IR spectroscopy to monitor temperature quantify cooling rates due to evaporation and radiation

\* More Speculative:

Photodynamics in pure and doped solid  $pH_2$  dopants based on  $p^+$  and  $e^-$  (i.e. generalizable to antimatter):  $e^-$  bubbles,  $H_3^+(H_2)_n$ ,  $H^-(H_2)_n$ , H,  $(oH_2)_2$  ...

UV, VIS, IR, microwave "fluorescence" efficiencies nonlinear optical processes:

stimulated Raman scattering (SRS) coherent anti-Stokes Raman scattering (CARS)

Optical refrigeration in solid pH<sub>2</sub>?!